Evaluation and continued development of aerosol-cloud interaction parameterizations for the GMI

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photo: S.Lance

Aerosol-Cloud Interaction Modules

Goal

Couple all aerosol-cloud-radiation interactions within a framework of parameterizations appropriate for global models.

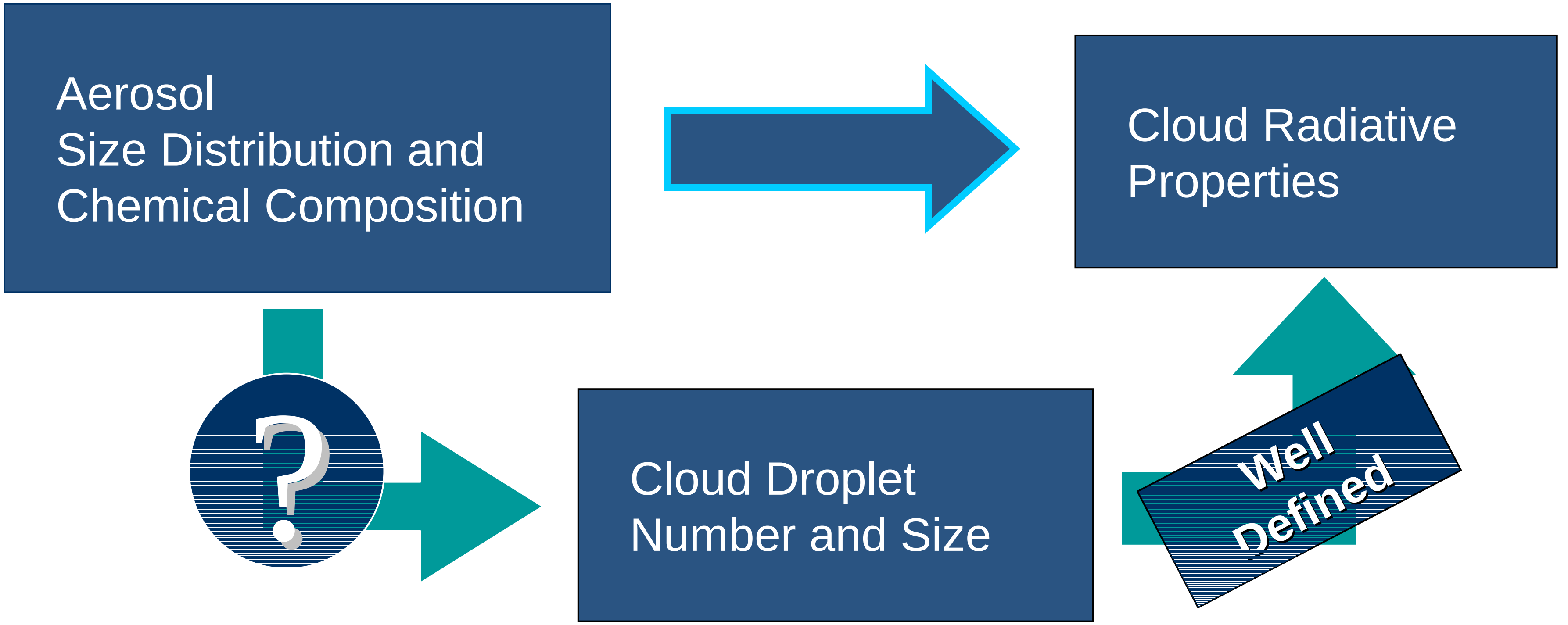
"Input" variables (from GCM)

- Cloud liquid water content.
- Aerosol size distribution and chemistry.
- Wind fields.
- Static stability/turbulence.

"Output" variables (to GCM)

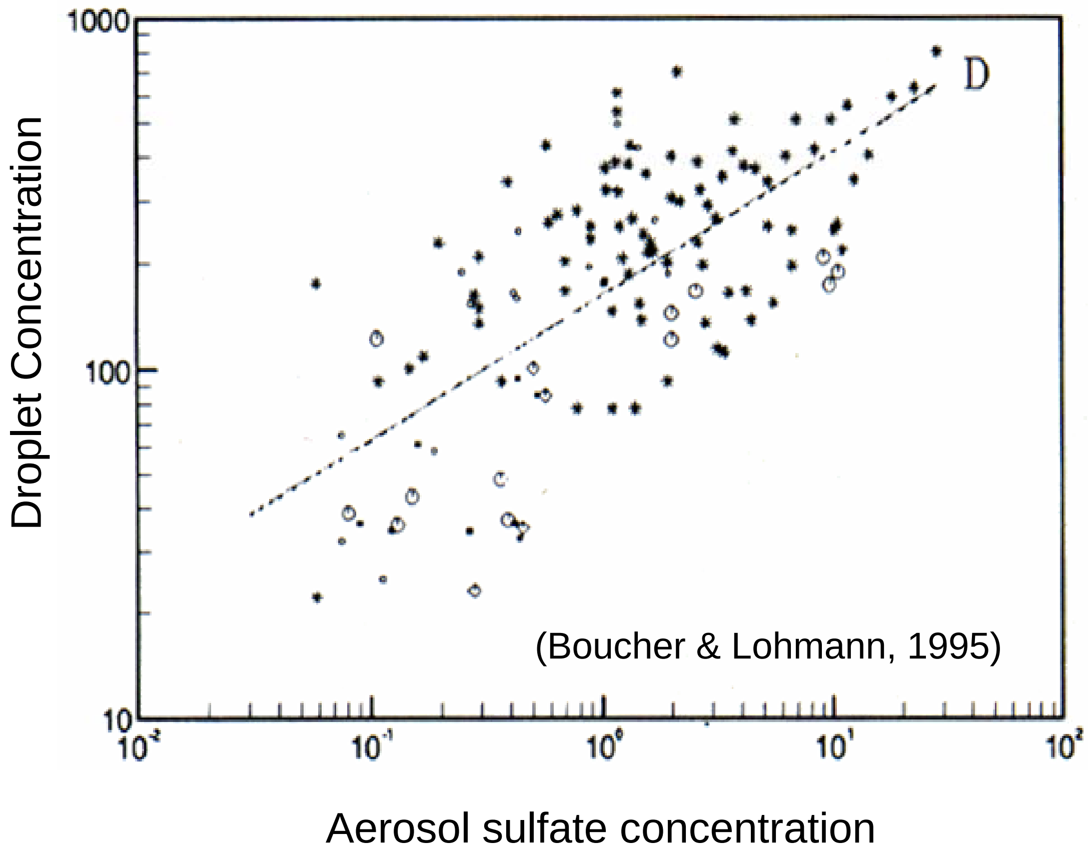
- Droplet number, distribution characteristics
- Cloud optical properties
- Cloud coverage, subgrid statistics

Quantification of the Indirect Effect



This problem has historically been reduced to finding the relationship between aerosol number concentration and cloud droplet number concentration. **Empirical** relationships are often used.

Simplest aerosol-cloud interaction module: correlations



Very large variability.

Why?

- Meteorology
- Cloud microphysics
- Chemical composition
- etc…

Pro: Very simple relationship to implement. Fast computation.

Con: Large predictive uncertainty, **without chance of improving**.

Organic Chemical Effects on Droplet Formation

- ## Surface Tension Depression

  Organics can decrease surface tension of droplets; this facilitates the ability of CCN to activate.

  Examples: succinic acid, humic substances

- ## Partial Solubility

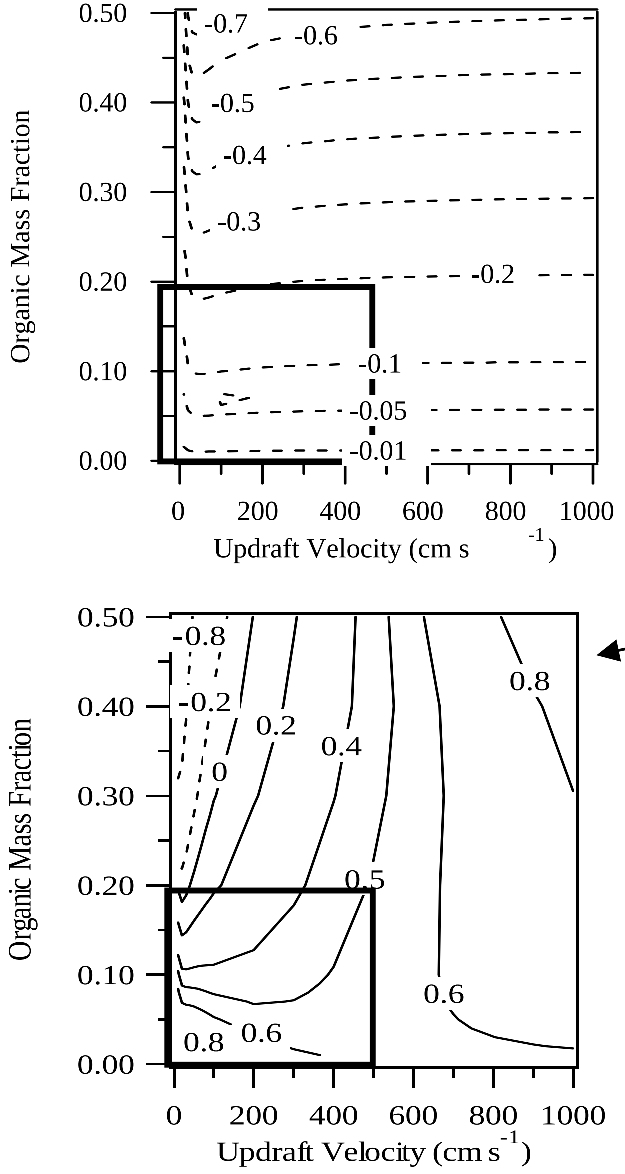
  Soluble or Partially Soluble organics lower the vapor pressure of CCN, facilitating their ability to activate.

- ## Changes in water vapor accommodation

  Organic Film-Forming Compounds can slow droplet growth by lowering the accommodation (i.e. “sticking”) coefficient.

  This can either facilitate or inhibit activation.

Chemical effects: an important source of variability



Sensitivity of N_d from chemical effects relative to updraft velocity for polluted aerosol

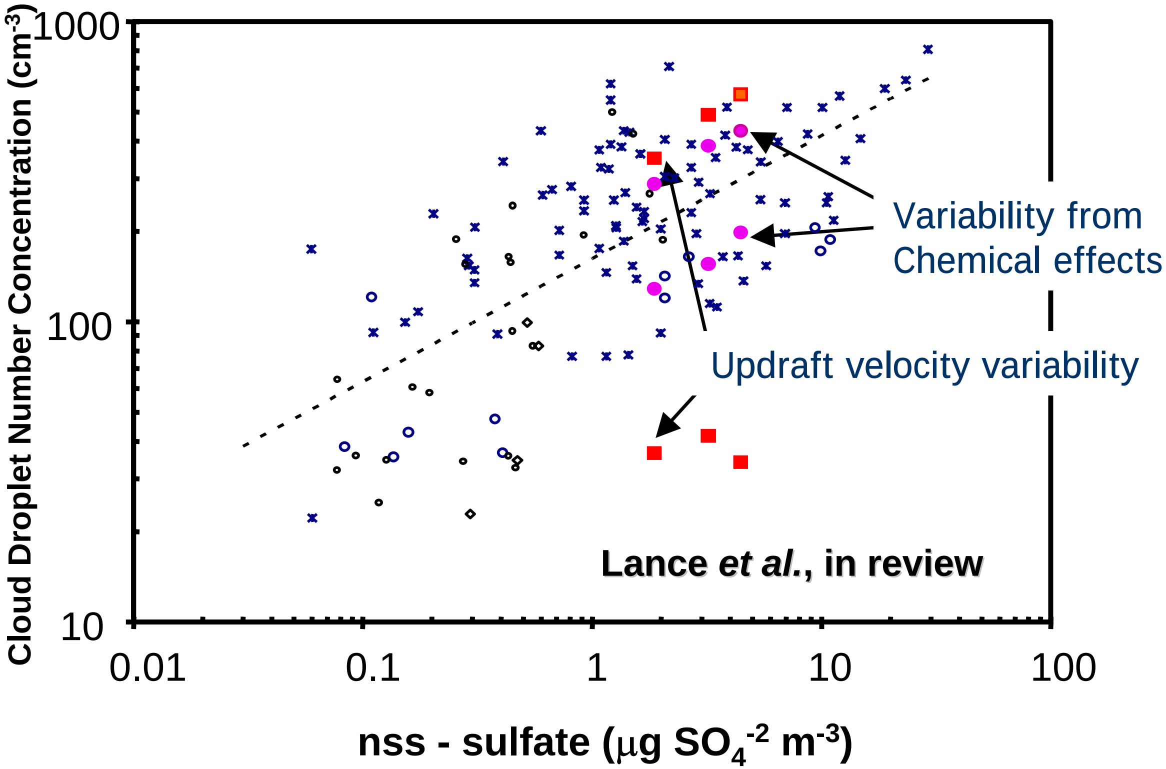
Without surface tension depression

With surface tension depression

Important:
Chemical effects not only affect N_d; they affect sensitivity to updraft velocity (spectrum).

Rissman et al., *JAS*, 2004

Chemical effects: an important source of variability



Variability from dynamic and chemical effects are significant and within the scatter.

This trend is **consistent** over a wide variety of aerosol types and conditions.

Traditional view: "Variations in cloud updraft velocity is believed to be one of the main reasons for the large variability in the observations with respect to the aerosol-cloud droplet empirical correlations" [IPCC, 2001].

Emerging (i.e., our) view: Aerosol chemical effects can be equally important. ***Both*** dynamics and chemistry must be considered in aerosol-cloud interaction studies.

Physically-based aerosol-cloud interaction modules

Uncertainties can be decreased by using first principles. Cloud droplet balance:

$$\frac{dN_{drop}}{dt} = Q_{activation} - Q_{evap} + Q_{advection} + \dots$$

$$\frac{d}{dt}\int_0^{\infty} p(w) N(w) dw$$

Probability of updraft *w*   Activated droplets for updraft *w*

Activation is the direct aerosol-cloud microphysical link. Two types of information are necessary for its calculation:

- Aerosol chemistry and size distribution (CCN)
- Representation of subgrid dynamics in cloud-forming regions.

Embedding a numerical activation model is too slow; must *parameterize*.

Mechanistic parameterizations: underlying ideas

Approach:

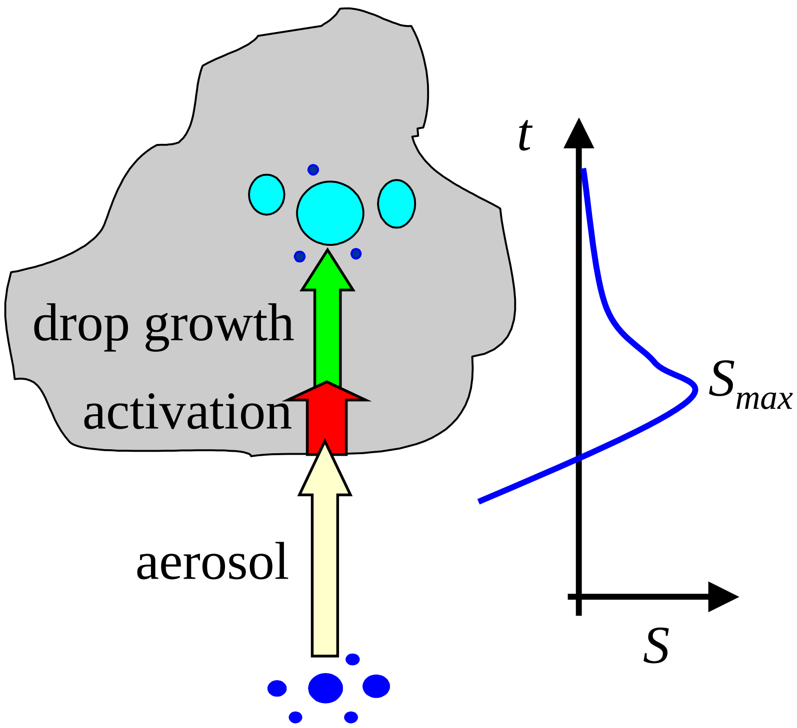
• Assume an aerosol size distribution and chemical composition below cloud.

• Aerosols rise into cloud.

• Expansion generates cooling and supersaturation.

• Aerosols activate into droplets.

• Köhler theory links aerosols to CCN properties.



Major challenge:

Derive expression for the condensational growth of CCN; include within the supersaturation balance for the parcel, and solve for the maximum.

Solution:

• Depends on the approach used in each parameterization.

• We use “Population splitting” (Nenes and Seinfeld, *JGR*, 2003)

Nenes and Seinfeld (2003) parameterization: Formulation

Input: P,T, updraft velocity (cooling rate), aerosol characteristics.
Output: Droplet number, S_{max}

How: Solve the algebraic equation for S_{max} (numerically) and N_d

Water vapor condensation from kinetically "limited" CCN

$$\frac{\pi}{2}\frac{\gamma\rho_w G S_{\max}}{aV}\left\{C_1\int_0^{S_{part}} f_1(s)\,ds + C_2\int_{S_{part}}^{S_{\max}} f_2(s)\,ds\right\} - 1 = 0$$

Water vapor condensation from CCN that "instantaneously" activate

Module Evaluation

How to evaluate?

Process-based approach: evaluate each component (process) of parameterizations using closure studies.

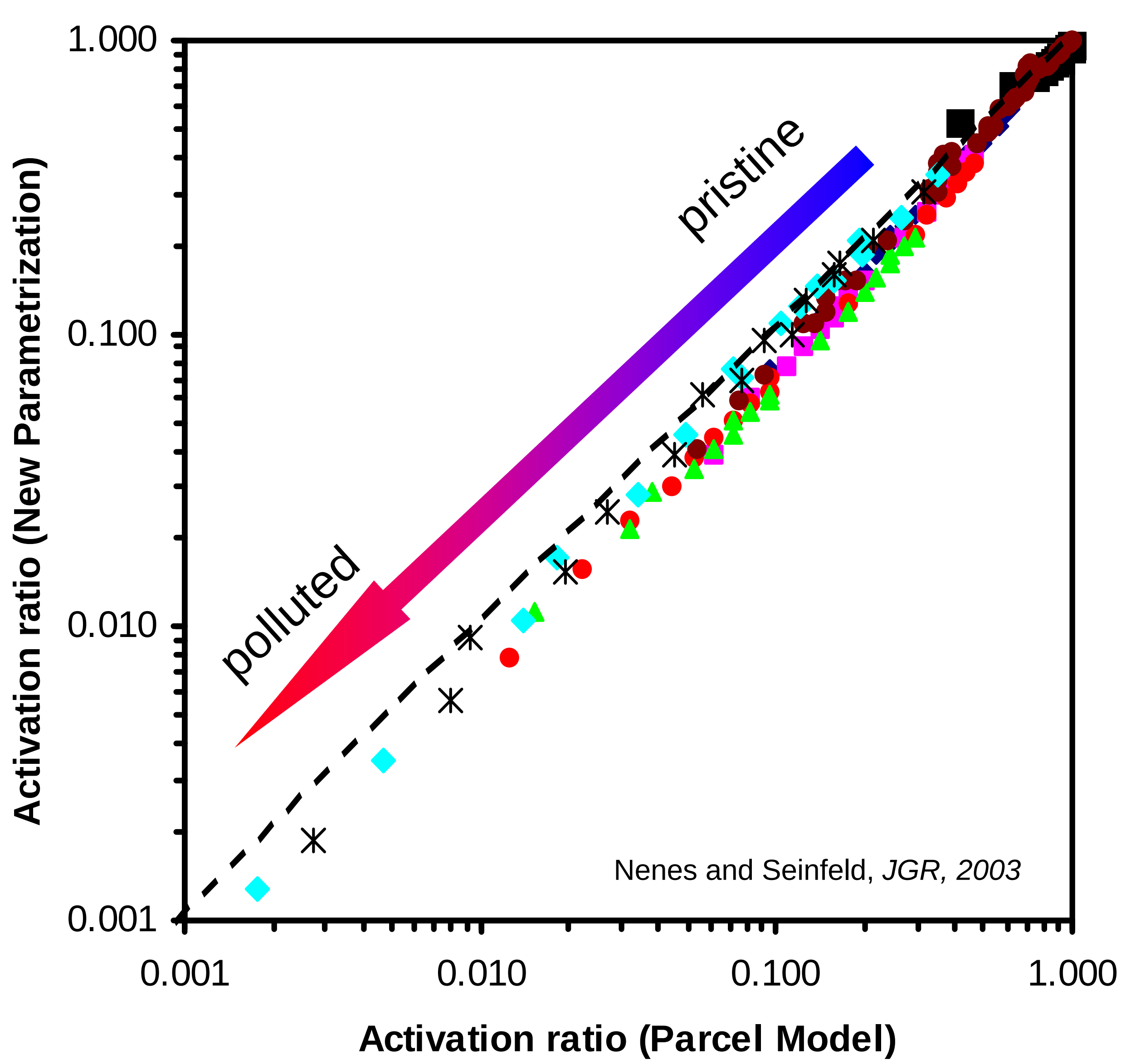
- Aerosol-CCN
- CCN-cloud droplet number
- Cloud droplet number – precipitation
- Chemistry-CCN evolution

Field data & detailed modeling must be used for all of evaluations. Comparison with models can suggest improvements; field data give the “reality check”.

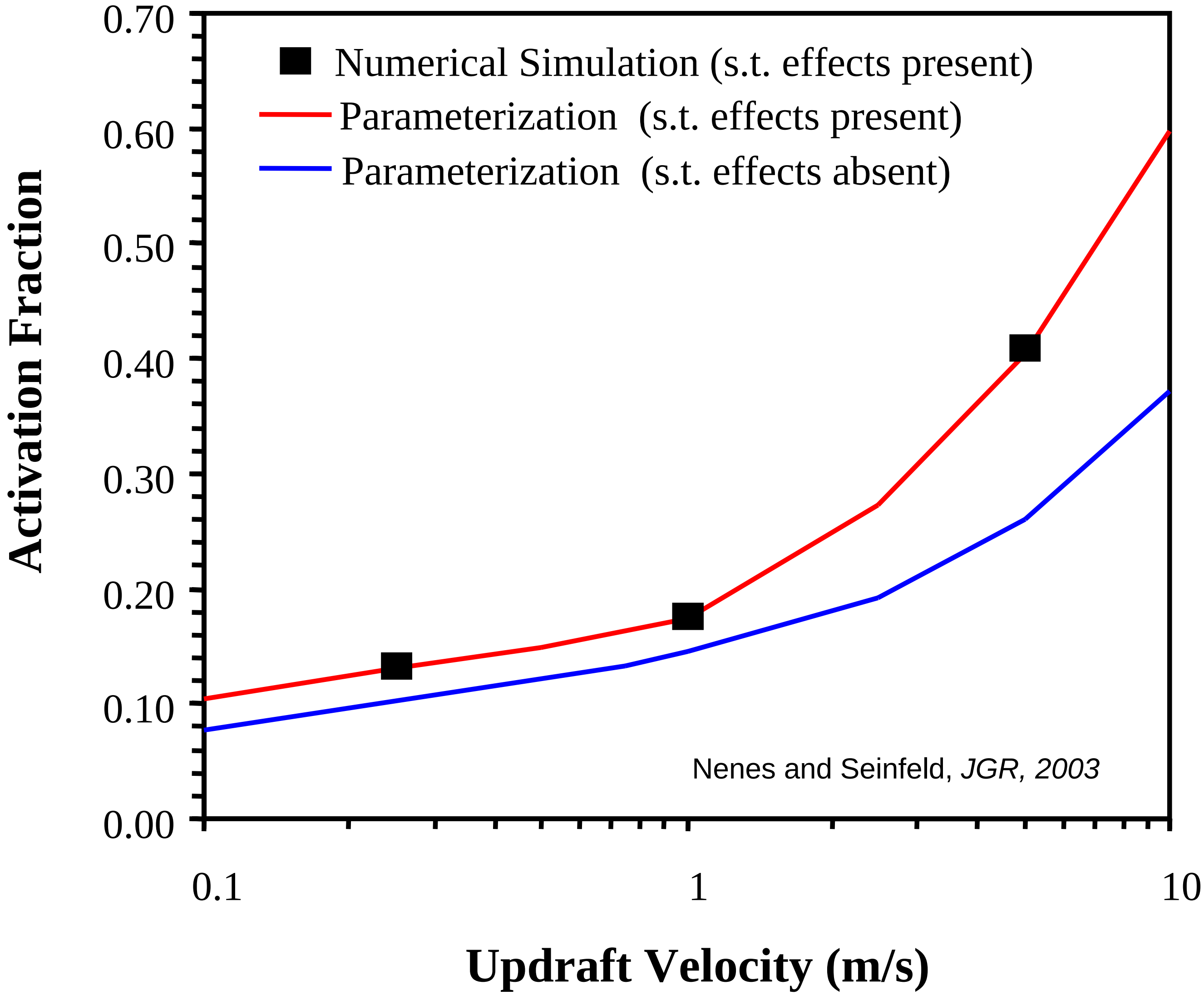
Coupling of multiple processes within simple modeling frameworks and Single Column Models to evaluate performance within GCM.

Point of concern for field data: the limited spatial/temporal scales covered.

N & S (2003) evaluation: compare with numerical model



N & S (2003) evaluation: compare with numerical model

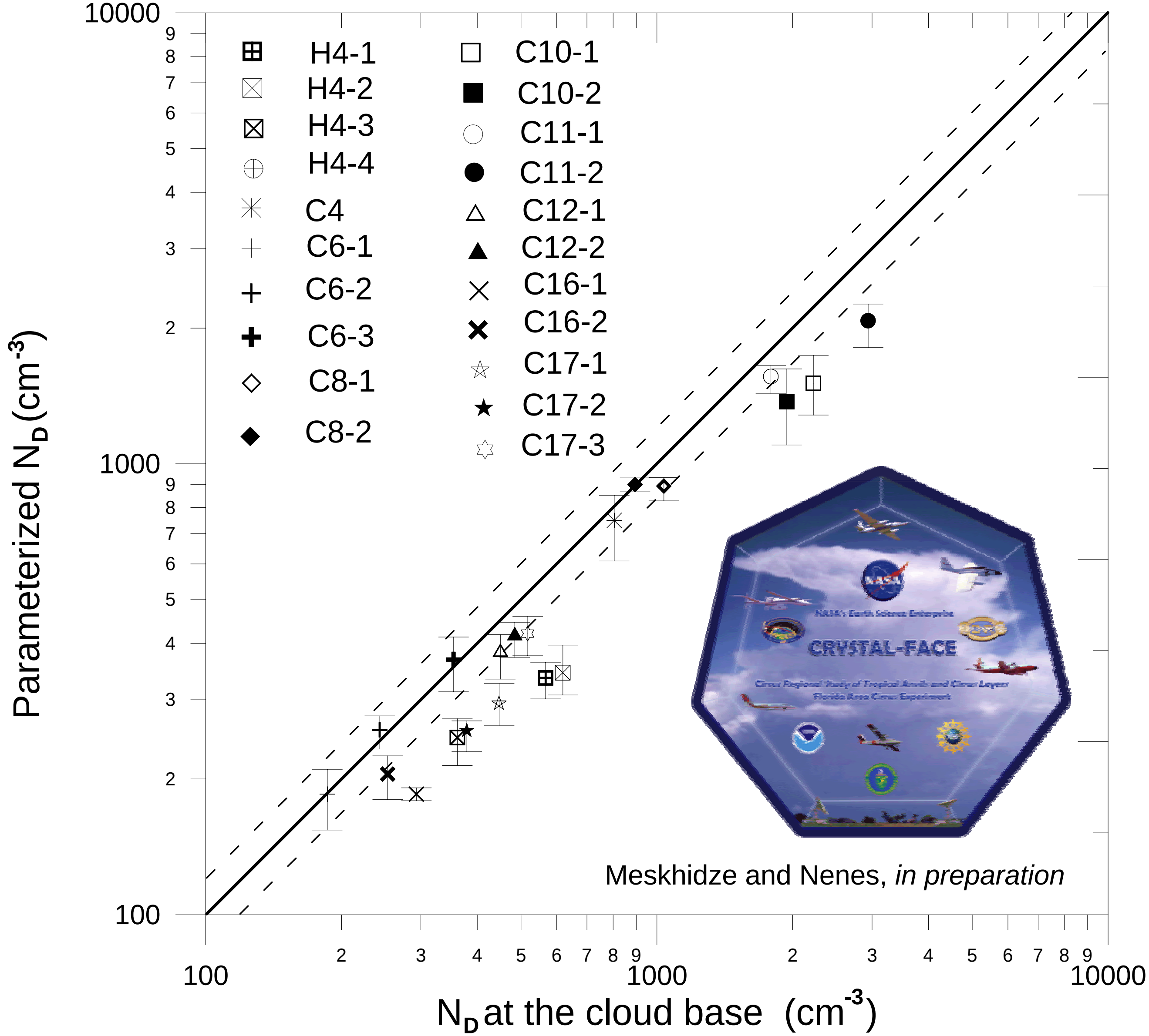


N & S (2003) evaluation: Field data comparison

Measure *in-situ* aerosol size/composition, updraft velocity and droplet concentration (CIRPAS Twin Otter).

Will the parameterization calculate the right number?

N & S (2003) evaluation: Field data comparison



Summary of parameterization evaluation

Pros:

• 10^3-10^4 times faster than full numerical model. Uses minimal amount of empirical info.

• Good agreement with detailed numerical model and field data.

• Chemically complex and externally mixed aerosol can be treated, including surface tension depression & partial solubility from organic species.

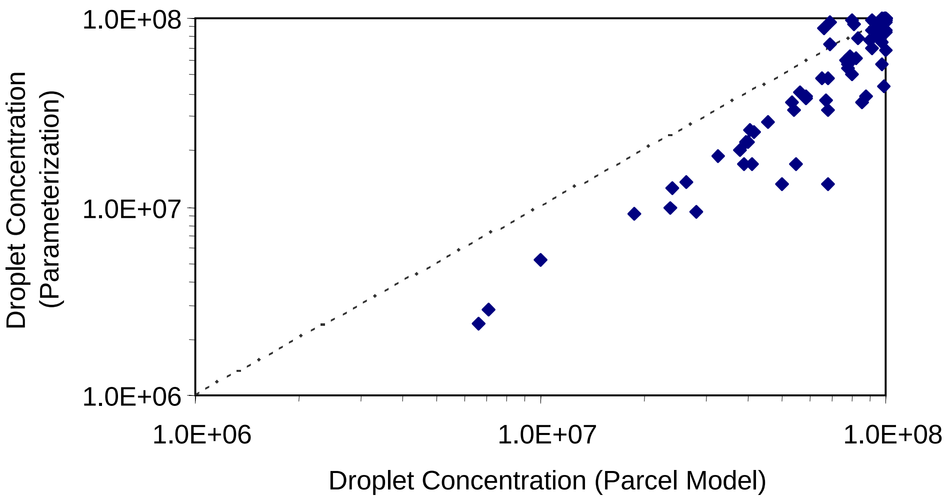
Cons:

• Tendency for **underestimation**, which increases as the water vapor accommodation coefficient decreases.

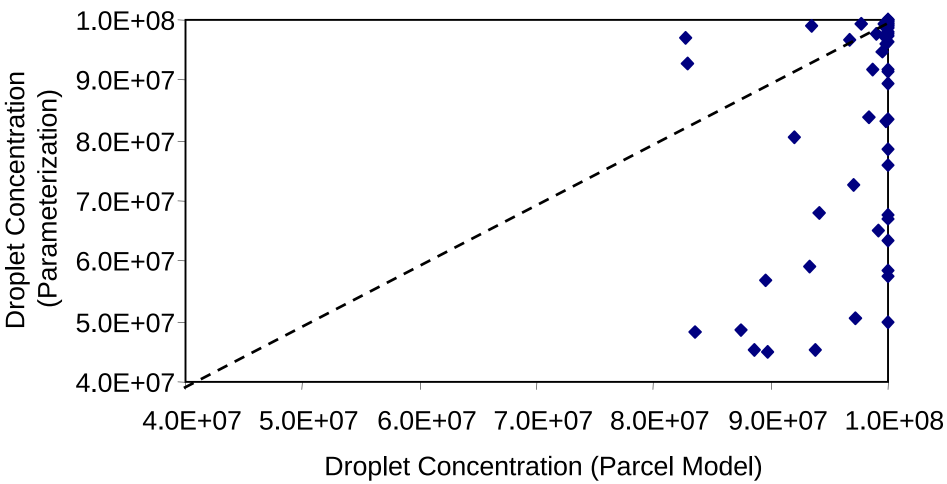
• Need to address to account for film forming compounds.

Note: Evaluations were for perfect accommodation of water vapor.

Underprediction: worstens as accommodation decreases



$\alpha = 0.042$



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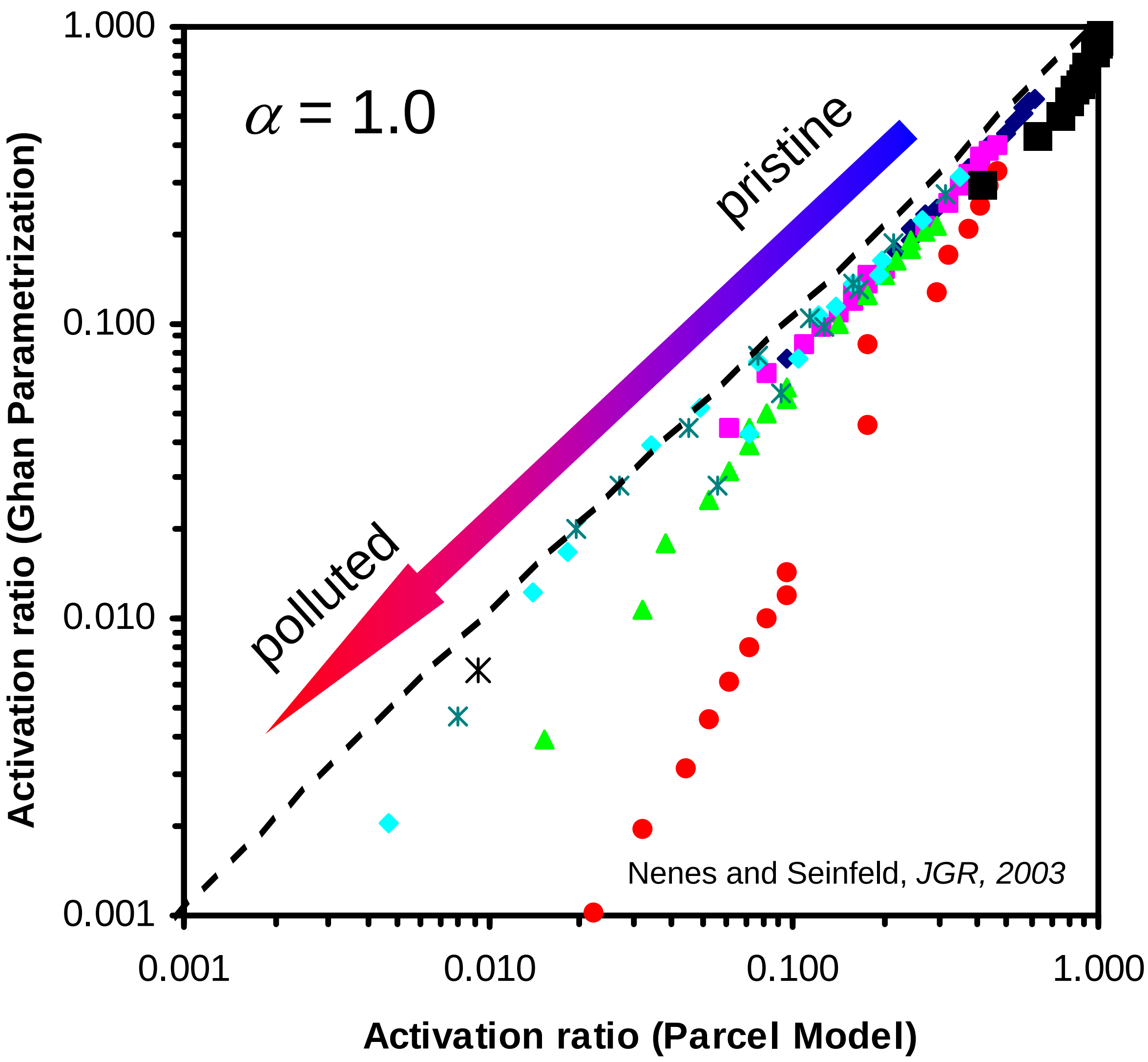
Fountoukis and Nenes, *in preparation*

Underprediction: common to many parameterizations

Abdul-Razzak *et al.* parameterization “family”



Source of problem: Water vapor diffusivity

Parameterizations neglect size effects on D_v

- Water vapor condenses too quickly on CCN.
- Supersaturation drops "faster" than it really should.
- Less CCN can thus activate.

$$D_v \qquad < \qquad D'_v = \frac{D_v}{1 + \frac{2D_v}{\alpha D_p}\sqrt{\frac{2\pi M_w}{RT}}}$$

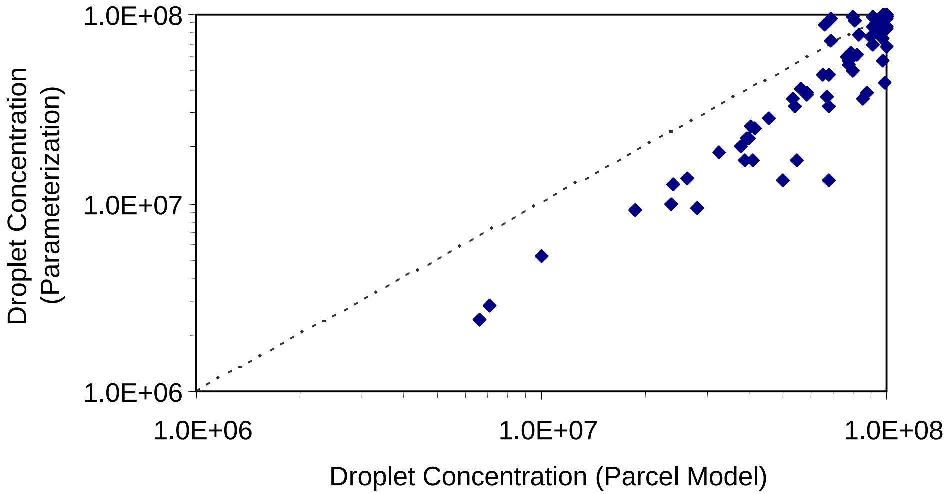
Solution(s):

- "Average" diffusivity for all CCN (fast, less accurate)
- Each section with its own diffusivity (slower, more accurate)

Note: Aerosol representation in model also determines approach

N&S (2003) parameterization evaluation with modified D_v

$\alpha = 0.042$



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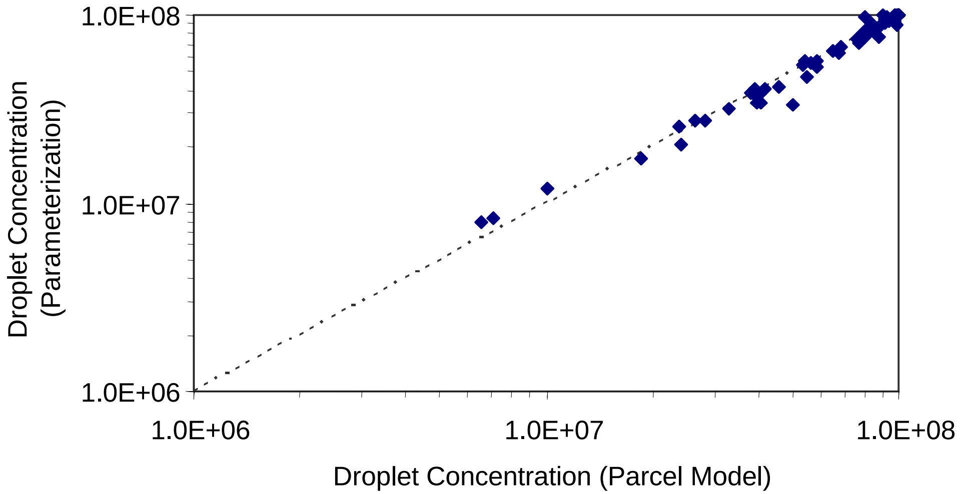

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Fountoukis and Nenes, *in preparation*

N&S (2003) parameterization evaluation with modified D_v

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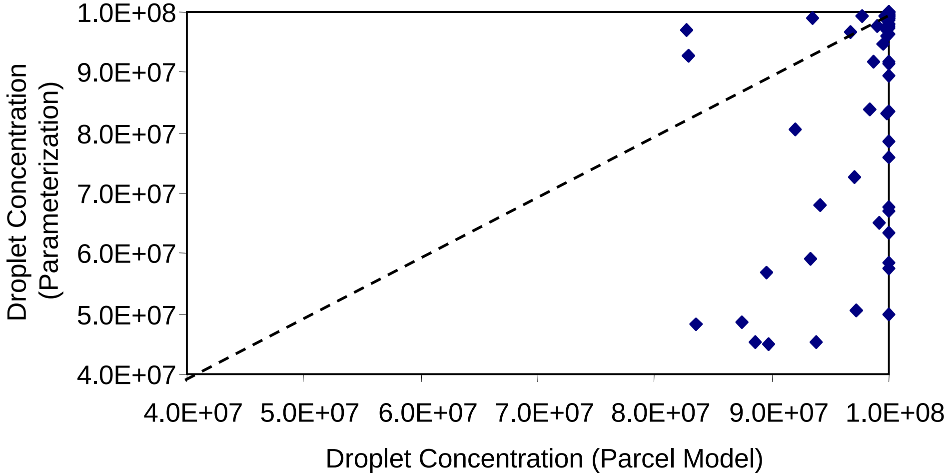

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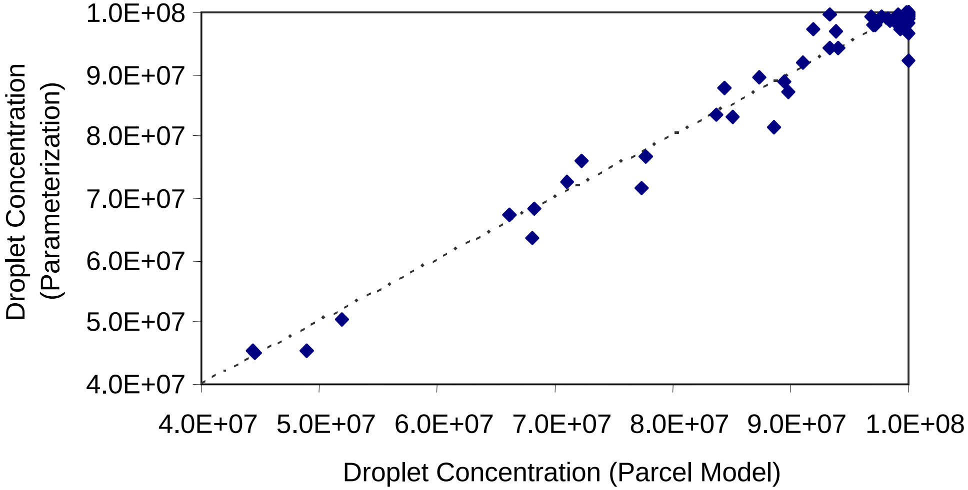

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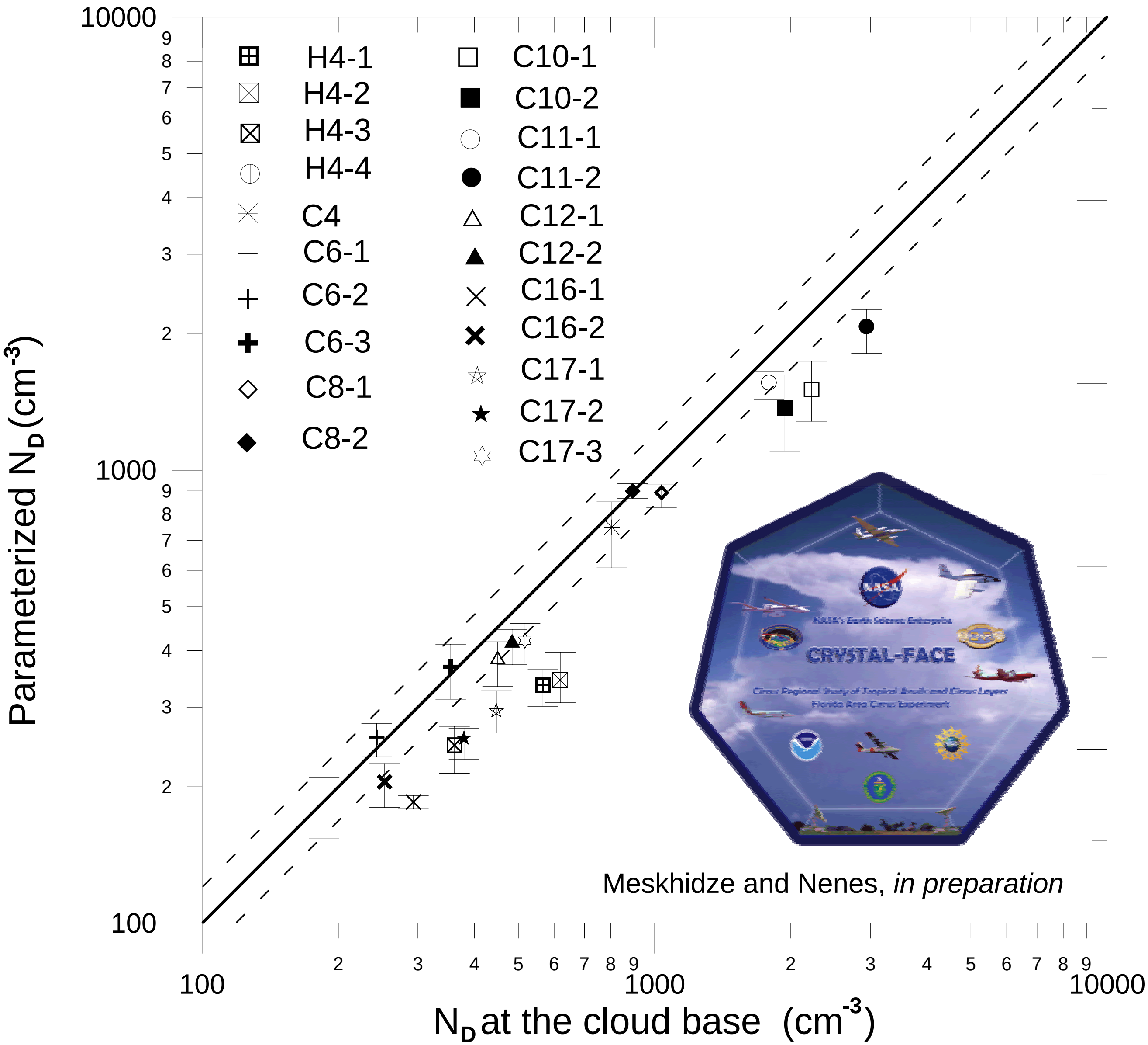

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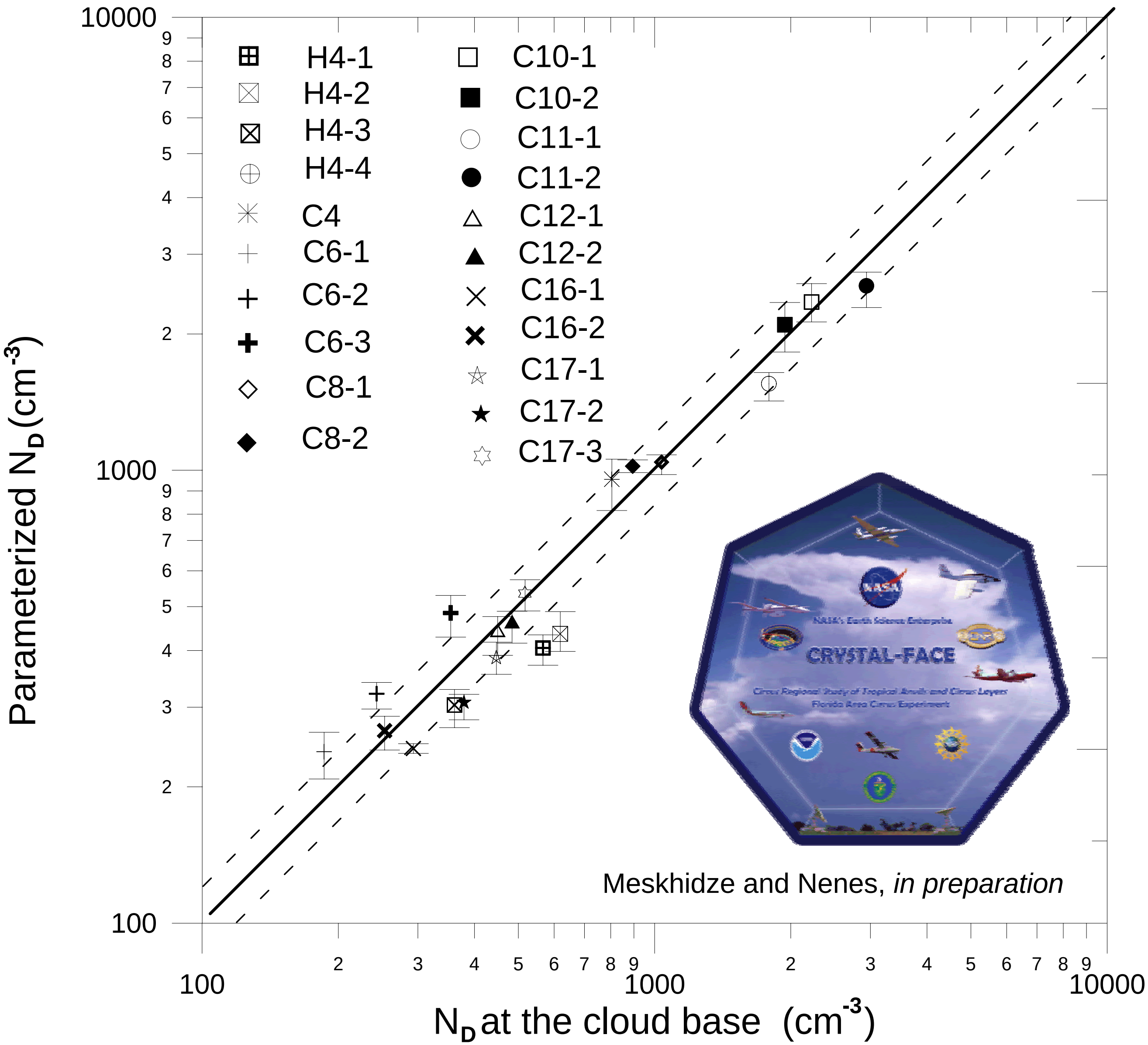
N&S (2003) parameterization evaluation with modified D_v



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Significant improvement!

Activation parameterization: active work

Accomplished:

- Significantly improved Nenes and Seinfeld (2003) by including size-dependence in the diffusivity.
- Can now consider CCN with film-forming compounds.
- Extended Abdul-Razzak et al. parameterizations to account for organic species.
- Knowledge from developing and improving Nenes and Seinfeld (2003) will be used to improve other parameterizations.

Currently working on:

- Include entrainment in parcels.
- Include variable updraft velocity.
- Deriving formulations for modal aerosol representation.
- Mixed phase clouds

Aerosol-cloud interactions in GMI: goals

Indirect forcing assessments

Perform a “traditional” sulfate (first) indirect forcing calculation and compare with published values.

Perform an indirect forcing calculation, where the contribution of anthropogenic aerosol to cloud optical depth (and its forcing) is assessed.

Explicitly test sensitivity of indirect forcing estimates to:

- Different size distribution representatons (e.g., modal vs. sectional)
- Aerosol mixing state
- Presence of a variety of “chemical” effects (organics, condensable gases and kinetically-limited aerosol)
- Size-dependant composition effect

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For more information and PDF reprints, http://nenes.eas.gatech.edu

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